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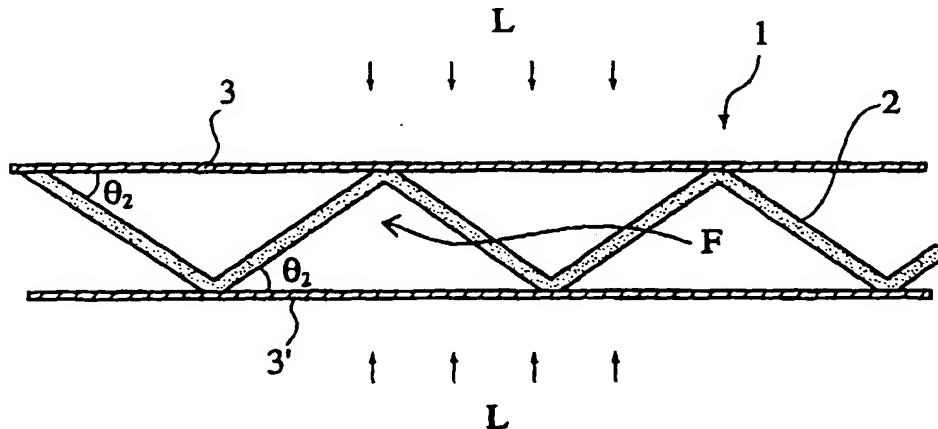
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### (54) Three-dimensional, photocatalytic filter apparatus

(57) A three-dimensional, photocatalytic filter apparatus is constituted by (a) a flow path means through which a fluid stream containing pollutants passes; (b) a plurality of porous filter plates each carrying a photocatalyst for decomposing the pollutants; (c) a light source; and (d) at least one support member for arranging each porous filter plate in a path of the fluid stream in the flow

path means, at such a three-dimensional position that each porous filter plate is inclined relative to both the fluid stream and the light source, to achieve sufficient contact with the pollutants and sufficient exposure to the light source.

Fig. 4



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**Description****BACKGROUND OF THE INVENTION**

[0001] The present invention relates to a three-dimensional, photocatalytic filter apparatus for removing organic or inorganic pollutants in a gas or liquid stream by using a semiconductor photocatalyst.

[0002] Ever increasing use of petrochemicals has caused serious complex pollution or contamination in a living environment. To solve the pollution and contamination problems, proposal has been made to provide a fluid purification method utilizing a photocatalytic oxidative decomposition of pollutants and pollutants by a semiconductor photocatalyst. For example, an apparatus or instrument having a porous substrate supporting a semiconductor photocatalyst is placed in a flow path of a fluid stream containing pollutants to bring the pollutants into contact with the semiconductor photocatalyst to decompose them.

[0003] In this method, to enhance the photocatalytic efficiency, it is needed to increase the photocatalytic surface area and to sufficiently activate the semiconductor photocatalyst with an electromagnetic wave having a wavelength capable of activating the photocatalyst. Several proposals have been made to provide a method for increasing a photocatalytic surface area, thereby increasing the opportunity of contact between the pollutants and the semiconductor photocatalyst, and a method for forming a photocatalyst layer. See Japanese Patent Laid-Open No. 5-309267, Japanese Patent Laid-Open No. 8-196903, etc.

[0004] Well known in the art is a method for forming a photocatalyst layer on a porous substrate such as a cluster of a structure having a large surface area (a cluster of sphere, etc.), a porous substrate (a non-woven fabric, a glass-wool, etc.), a spongy substrate, so as to increase the photocatalytic surface area.

[0005] In this method, the porous substrate often absorbs or shuts out an activating light, thereby failing to activate the semiconductor photocatalyst deposited on an inner part of the porous substrate. When a cluster made of a silica glass, a soda glass, etc. or a glass wool is used as the porous substrate, the activating light reaches comparatively deeper inside. However, a diameter of the porous substrate is limited to 10-30 mm even in this case. The porous substrate having large light-shielding properties has a limited thickness of about 2 mm or less. Therefore, this method cannot be widely used in various industrial applications.

[0006] Some proposals also have been made to provide a shape of a photocatalytic filter and an arrangement of a photocatalytic filter. For example, Japanese Patent Laid-Open No. 7-108138 discloses a photocatalytic filter apparatus in which photocatalyst-supporting slats are arranged in the form of a blind in a flow path of a fluid stream. Japanese Patent Laid-Open No. 8-121827 discloses a photocatalytic filter apparatus

in which a photocatalyst-supporting, non-woven fabric bent in an angle bar shape is placed at the front and back of an ultraviolet lamp installed in a flow path of a fluid. Japanese Patent Laid-Open No. 9-187491 discloses a photocatalytic filter apparatus placed in a flow path of a fluid, the apparatus comprising a plurality of porous substrates each supporting a photocatalyst on both surfaces and radially fixed around a light source. Japanese Patent Laid-Open No. 9-248426 discloses a movable, photocatalytic filter apparatus comprising a ultraviolet source placed inside a photocatalyst-supporting, convex porous substrate for conveying or stirring a fluid.

[0007] Pollutants contacting with the semiconductor photocatalyst are oxidatively decomposed by activated radical or super oxide anion such as  ${}^1\text{O}_2$ ,  $\cdot\text{OH}$ , etc. If a large amount of pollutants were tried to be decomposed by the conventional photocatalytic filter apparatus mentioned above, the pollutants would not fully come into contact with the photocatalyst, suffering disadvantages that it takes a lot of time to ensure contact of all the pollutants with the photocatalyst. It is difficult to increase permeability in the conventional photocatalytic filter apparatuses, while enhancing decomposition yield. Additionally, to enhance photocatalytic efficiency, a suitable activating light should sufficiently illuminate the semiconductor photocatalyst. Therefore, the development of a photocatalytic filter apparatus capable of efficiently illuminating the semiconductor photocatalyst has been desired.

[0008] In the oxidative decomposition of nitrogen compounds, sulfur compounds, chlorine compounds, etc., intermediate products tend to adhere to a surface of the semiconductor photocatalyst. Because the attached intermediate products should be removed from the semiconductor photocatalyst at regular intervals, the photocatalytic filter apparatus is required to have a structure capable of being easily cleaned to prevent reducing a decomposition-capacity thereof.

**OBJECT AND SUMMARY OF INVENTION**

[0009] Accordingly, the first object of the present invention is to provide a three-dimensional, photocatalytic filter apparatus having an increased photocatalytic surface area, thereby increasing contact between pollutants and a semiconductor photocatalyst.

[0010] The second object of the present invention is to provide a three-dimensional, photocatalytic filter apparatus ensuring adequate irradiation on the entire photocatalytic surface, and making good use of scattered light.

[0011] The three-dimensional, photocatalytic filter apparatus and the three-dimensional, photocatalytic filter apparatus preferably have such a structure that impurities such as intermediate products, etc. attached thereto are easily removed.

[0012] As a result of intense research in view of the

above objects, the inventors have found the following facts:

(1) When a plurality of porous filter plates are three-dimensionally assembled in such a manner that each filter plate is inclined relative to the fluid stream, sufficient contact is achieved between the fluid stream and the semiconductor photocatalyst, thereby removing the intermediate products efficiently.

(2) Sufficient irradiation on the photocatalytic surface and enough utilization of scattered light are ensured:

(a) when at least one three-dimensional, porous filter plate is arranged around a light source;

(b) when a plurality of inclined, porous filter plates are three-dimensionally assembled around a light source and axially placed in a pipe through which contaminant-containing fluid flows, in such a manner that a surface of each filter plate is sufficiently exposed to the light, and that sufficient contact is achieved between the fluid stream and the semiconductor photocatalyst;

(c) when a porous filter plate providing a sufficient contact of a fluid stream with a semiconductor photocatalyst, constituted by a porous substrate and a porous layer made of a metal material and a photocatalyst layer; a support member equipped with a light source therein; and a pipe in which a fluid stream flows in an axial direction, the porous substrate being a wire cloth of plain weave, the porous layer being sintered-metal layer disposed on the porous substrate, the porous filter plate and the support member being placed in the pipe, the porous filter plate being fixed around the support member spirally.

[0013] A component constituted by the support member and the photocatalytic filter will be called "three-dimensional, photocatalytic filter unit" below.

[0014] The above objects are met by the three-dimensional photocatalytic filter apparatus defined in the claims.

#### BRIEF DESCRIPTION OF THE DRAWINGS

[0015]

Fig. 1 (a) is a schematic, partial, cross-sectional view showing one example of a porous filter plate comprising a porous substrate and a porous layer according to the present invention;

Fig. 1 (b) is a schematic, partial, cross-sectional view showing another example of a porous filter

plate comprising a porous substrate and a porous layer according to the present invention;

Fig. 2 is a schematic perspective view showing a three-dimensional, photocatalytic filter apparatus according to one embodiment of the present invention;

Fig. 3 is a cross-sectional view showing the same three-dimensional, photocatalytic filter apparatus as in Fig. 2;

Fig. 4 is a cross-sectional view showing a three-dimensional, photocatalytic filter apparatus according to another embodiment of the present invention; Fig. 5 (a) is a front view showing a three-dimensional, photocatalytic filter apparatus according to a further embodiment of the present invention;

Fig. 5 (b) is a cross-sectional view taken along the line A-A in Fig. 5 (a);

Fig. 6 is a front view together with a plan view and a side view each showing a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention;

Fig. 7 is a front view showing a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention;

Fig. 8 is a side view showing of a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention;

Fig. 9 is a cross-sectional view showing a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention;

Fig. 10 is a perspective view showing a three-dimensional, photocatalytic filter element in the three-dimensional, photocatalytic filter apparatus of Fig. 9;

Fig. 11 is a front view showing one type of a support member in the three-dimensional, photocatalytic filter element of Fig. 10;

Fig. 12 is a schematic view showing the structure of the three-dimensional, photocatalytic filter unit of Fig. 10;

Fig. 13 is a front view showing another type of a support member for a three-dimensional, photocatalytic filter unit;

Fig. 14 is a perspective view showing a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention; and

Fig. 15 is a partial, cross-sectional view showing a three-dimensional, photocatalytic filter apparatus according to a still further embodiment of the present invention.

DETAILED DESCRIPTION OF THE PREFERRED EMBODIMENTS

[1] Porous filter plate

[0016] A porous filter plate for the three-dimensional, photocatalytic filter apparatus of the present invention is constituted by a porous substrate, a porous layer composed of fine particles fixed onto at least one surface of the porous substrate, and a photocatalyst layer deposited on the surface of the porous layer.

(A) Porous substrate

[0017] As shown in Figs. 1 (a) and (b), a porous substrate 20 of each filter plate 2 has a lot of pores 21 through which a fluid (gas or liquid) containing pollutants passes. Fine particles 22 are deposited onto a surface of the porous substrate 20 to form a porous layer 23 to such an extent that the pores 21 are sufficiently covered by the porous layer 23 of fine particles 22.

[0018] In Figs. 1 (a) and (b), "S" represents a surface area of a region of the porous substrate 20 that is exposed to an activating light, "a" represents amount of a fluid stream F passing through the porous substrate 20 per a unit time, "b" represents a surface area of a region of the porous layer 23 between adjacent pores 21, and "c" represents a surface area of the side of the porous substrate 20. Assuming that the filter plate 2 is irradiated with the activating light L from both sides, from above and below in Fig. 1, the porous substrate 20 preferably has such a structure as to provide a maximum ratio b/S and a minimum ratio of c/S. Accordingly, the structure shown in Fig. 1 (a) is more preferable than that shown in Fig. 1 (b).

[0019] The porous substrate 20 may be constituted by a net or mesh of wires, a perforated sheet, etc. in this case, the wire and the perforated sheet are preferably made of metals such as Fe or its alloys such as austenitic stainless steel (SUS304, SUS310, SUS316, etc.); Al or its alloys such as Al-Si-Mg alloy; Ti or its alloys such as Ti-Mn alloy and Ti-Cr alloy; and Cu or its alloys. When Fe or Fe alloy is used for the porous substrate 20, a non-oxydizing film such as iron oxide is preferably formed on a surface of the porous substrate 20. The porous substrate 20 may also be constituted by a porous ceramic, glass, metal or plastic sheet.

(B) Porous layer

[0020] The porous layer 23 is formed on a surface of the porous substrate 20 to increase a surface area thereof. The porous layer 23 may be composed of fine particles 22 of ceramics, glass, metals, plastics, etc. The fine particles 22 are preferably Fe or its alloys such as austenitic stainless steel (SUS304, SUS310, SUS316, etc.); Al or its alloys such as Al-Si-Mg alloy; Ti or its alloys such as Ti-Mn alloy and Ti-Cr alloy; and Cu or its

alloys. The materials for the fine particles 22 are preferably the same as those of the porous substrate 20.

[0021] Fine particles having extremely small sizes are expensive, while those having too large sizes fail to provide the porous layer 23 with a sufficient surface area. Accordingly, the average diameter of the fine particles 22 is preferably 10-400  $\mu\text{m}$ . The shapes of the fine particles 22 are not strictly limited, and they may have regular shapes such as sphere and granule or irregular shapes with sharp ends as in angular particles. The fine particles 22 may also be flaky.

[0022] To integrally fuse the fine particles 22 to the porous substrate 20, the fine particles 22 and the porous substrate 20 are preferably made of the same metals. When different materials are used, it is preferable to use suitable binders to securely adhere the fine particles 22 to the porous substrate 20. In this case, the different materials are preferably combined such that their linear thermal expansion coefficients are substantially equal or near to each other, or one of the materials is preferably easily stretchable to absorb the thermal expansion of the other material. The binder may be inorganic glass, frit, metal powder, thermoplastic resins, etc.

[0023] The porous layer 23 may be formed on the porous substrate 20 by applying a slurry of fine particles 22 in a liquid medium such as water, alcohol or a mixture thereof (solid content: 60-80 weight %) to a surface of the porous substrate 20, drying the resultant coating, and sintering the fine particles 22.

[0024] The fine particles 22 are preferably applied onto the porous substrate 20 by dipping, spraying, screen-printing, etc. The application of the fine particles 22 may be carried out several times.

[0025] The sintering temperature of the fine particles 22 may be selected depending on a material thereof. When the sintering temperature is too low, the resultant porous substrate 23 does not have a sufficient sintering density, resulting in poor mechanical strength. On the other hand, when the sintering temperature is equal to or higher than the melting point of the fine particles 22, the fine particles 22 are melted, resulting in coarse pores in the porous substrate 23. Specifically, the sintering temperature is preferably 800-1000  $^{\circ}\text{C}$  for SUS, Ti alloys and Cu alloys, and 300-400  $^{\circ}\text{C}$  for aluminum alloys.

[0026] The porous layer 23 has a pore size preferably in the range of 5-1000  $\mu\text{m}$ . If the pore size of the porous layer 23 were more than 1000  $\mu\text{m}$ , dust would not be filtered out. When the thickness of the porous layer 23 is less than 10  $\mu\text{m}$ , the porous layer 23 has insufficient mechanical strength. On the other hand, when the thickness of the porous layer 23 exceeds 100  $\mu\text{m}$ , the porous layer 23 is highly resistant to a fluid flow, causing excessive pressure loss. Therefore, the thickness of the porous layer 23 is preferably 10-100  $\mu\text{m}$ .

## (C) Photocatalyst layer

[0027] Semiconductor photocatalysts usable in the present invention include  $TiO_2$ ,  $ZnO$ ,  $SrTiO_3$ ,  $CdS$ ,  $CdO$ ,  $CaP$ ,  $InP$ ,  $In_2O_3$ ,  $CaAs$ ,  $BaTiO_3$ ,  $K_2NbO_3$ ,  $Fe_2O_3$ ,  $Ta_2O_5$ ,  $WO_3$ ,  $SnO_2$ ,  $Bi_2O_3$ ,  $NiO$ ,  $Cu_2O$ ,  $SiC$ ,  $SiO_2$ ,  $MoS_2$ ,  $MoS_3$ ,  $InPb$ ,  $RuO_2$ ,  $CeO_2$ , etc. Most preferable among them is anatase-type  $TiO_2$  because of low cost, high stability and harmlessness to human body.

[0028] The semiconductor photocatalyst may be coated on the porous layer 23 by spraying, dipping, etc. of a sol of a semiconductor photocatalyst such as  $TiO_2$ . After coating and drying, the semiconductor photocatalyst is securely bound to the fine particles 22 by baking preferably at 50-500 °C.

[0029] When the semiconductor photocatalyst sol contains amorphous peroxytitanic acid or titanium oxide at a dry weight ratio of 1:1 to 1:5 on the basis of a titanium element to the semiconductor photocatalyst, the semiconductor photocatalyst is securely deposited on the fine particles 22 at relatively low temperatures. Further, small amounts of Pt, Ag, Rh,  $RuO_2$ , Nb, Cu, Sn,  $NiO$ , etc. may be added to provide antibacterial properties and mildew resistance to the resultant photocatalyst layer. Also, to enhance the photocatalytic oxidative/reductive decomposition of pollutants in the fluid stream F, inorganic substances such as zeolite, silica, alumina, zinc oxide, magnesium oxide, rutile-type titanium oxide, zirconium phosphate, etc., activated carbon, porous phenol resins, and porous melamine resins may be added.

[0030] A primer layer may preferably be formed on the porous layer 23 by spraying a hydrosol of titanium peroxide, etc. before forming the semiconductor photocatalyst layer on the fine particles 22. The titanium peroxide primer layer provides the fine particles 22 with enough wettability, so that the semiconductor photocatalyst sol is evenly distributed on the fine particles 22.

[0031] The titanium peroxide primer layer is also effective for the porous substrate 20 and the fine particles 22 both made of a metal such as stainless steel, because they are made well wettable with the semiconductor photocatalyst sol. Titanium peroxide also serves as a binder. Since titanium peroxide contains no ceramic components, it has high affinity for a metal, ensuring the semiconductor photocatalyst to securely adhere to the fine metal particles 22 via titanium peroxide. As a result, the photocatalyst would not be detached from the porous layer 23, even when the filter plate 2 is deflected or vibrated.

## (D) Application of porous filter plate

[0032] The porous filter plate may be formed in various shapes depending on its use, and it may be curved or twisted, if necessary. In one embodiment, the photocatalyst layer may be formed on both sides of the porous substrate 20. In this case, light sources are pref-

erably placed on both sides of the porous filter plate 2 so that both surfaces of the porous filter plates 2 are irradiated at the same time.

[0033] Since the porous filter plate 2 comprises the porous substrate 20, it may serve not only as a filter element but also as a porous partition. For example, the porous filter plate 2 may be utilized for cleaning the air in a closed chamber or room. When a part of inner wall of a closed chamber of a refrigerator, etc. is formed with a porous, photocatalytic filter plate, and when the air is circulated in the closed chamber through the porous filter plate, undesirable substances such as ethylene, which ages vegetables and fruits, and smell substances such as hydrogen sulfide, mercaptan, etc. can be removed from the air. Since the inner wall serves as a filter, no additional filter is needed, permitting the maximum use of an internal space of the chamber.

[0034] In addition to the fluid-cleaning functions, the porous, photocatalytic filter plate exhibits a sound-absorbing function, a vision-shielding function, a defoaming function, and a wave-absorbing function.

[0035] When the porous, photocatalytic filter plate is used as a sound barrier between a sidewalk and a roadway, the sound-absorbing effect is obtained in addition to the photocatalytic decomposition of pollutants such as  $NO_x$  and  $SO_x$  in the air. The traffic noise is reduced because the propagation energy of sound wave is absorbed by the porous, photocatalytic filter plate while passing through the complicated porous structure thereof. The photocatalytic filter exhibiting the shielding effect may be used as a blind, a partition, etc.

[0036] The wave motion of a liquid stream brings pollutants to uneven contact with the porous, photocatalytic filter plate 2. However, the liquid stream passing through the porous, photocatalytic filter plate 2 ensures uniform contact of pollutants with the photocatalyst. Bubbles on a surface of the liquid stream may cover the photocatalytic surface, preventing enough contact between the pollutants and the photocatalytic surface.

[0037] As the liquid stream penetrates into the porous, photocatalytic filter plate 2, bubbles are divided to finer ones due to the complicated porous structure of the filter plate 2, thereby preventing the photocatalytic surface from being covered by the bubbles.

[0038] The porous filter plate 2 may also be used as a filter for an air-conditioning system, a filter for a waste gas-treating apparatus, a building component such as wallboard, a wall material for algae-free aquarium and swimming pool, etc.

## [2] Three-dimensional, photocatalytic filter apparatus

[0039] In one embodiment shown in Figs. 2 and 3, the three-dimensional, photocatalytic filter apparatus 1 comprises a plurality of porous filter plates 2 inclined in the same direction at an angle of  $\theta_1$  relative to the direction of the fluid stream F and arranged at a regular interval in a longitudinal direction, and flow path means 3, 3'

for defining a flow path of the fluid stream F. Both ends of each porous filter plate 2 are supported by support members (not shown). The fluid stream F flows through a plurality of the inclined porous filter plates 2. Light sources are positioned on both sides of the flow path means 3, 3' such that both surfaces of each porous filter plate 2 is exposed to an activating light L. The inclination angle  $\theta_1$  is preferably 15-75°, more preferably 30-60°, particularly 45°.

[0039] When pollutants such as nitrogen compounds, sulfur compounds, chlorine compounds, etc. are decomposed by the photocatalyst carried by the porous filter plate 2, intermediate products formed during their decomposition reaction tend to attach to the surface of the semiconductor photocatalyst. Because the intermediate products should be removed regularly, the three-dimensional, photocatalytic filter apparatus should have a structure permitting easy cleaning of the porous filter plates 2 to maintain their decomposition capacity. Thus, the three-dimensional, photocatalytic filter apparatus preferably has such a structure that water, weak acidic water, etc. can easily be injected to remove the intermediate products.

[0040] In another embodiment shown in Fig. 4, a porous filter plate 2 is corrugated at an angle of  $\theta_2$  relative to the direction of the fluid stream F, and light sources are placed on both sides of flow path means 3, 3'. The corrugating angle  $\theta_2$  is preferably 15-75°, more preferably 30-60°, particularly 45°.

[0041] A three-dimensional, photocatalytic filter apparatus shown in Figs. 5 (a) and (b) has a honeycomb structure constituted by two groups of porous filter plates 2a, 2b. Each porous filter plate 2a in one group is inclined by an angle  $\theta_3$  relative to the fluid stream F (see Fig. 5 (a)), and rotated around its axis by an angle  $\theta_5$  (see Fig. 5 (b)). Likewise, each porous filter plate 2b in the other group is inclined in the opposite direction by an angle  $\theta_4$  relative to the fluid stream F (see Fig. 5 (a)), and rotated around its axis by an angle  $\theta_6$  (see Fig. 5 (b)). Each of the inclination angles  $\theta_3$  and  $\theta_4$  is preferably 15-75°, more preferably 30-60°, particularly 45°. Also, each of the rotation angles  $\theta_5$  and  $\theta_6$  is preferably 15-75°, more preferably 30-60°, particularly 45°. With such arrangement of the porous filter plates 2a, 2b, both surfaces of each porous filter plate 2a, 2b are sufficiently exposed to an activating light L.

[0042] Fig. 6 shows a three-dimensional, photocatalytic filter apparatus according to a further embodiment having two groups of porous filter plates 2c, 2d assembled in a lattice structure. Light sources (not shown) are disposed on both sides of the porous filter plates 2c, 2d. The porous filter plates 2c in one group extend vertically and are rotated around their axes by an angle  $\theta_7$ . The porous filter plates 2d in the other group extend horizontally and are rotated around their axes by an angle  $\theta_8$ . Each of the inclination angles  $\theta_7$  and  $\theta_8$  is preferably 15-75°, more preferably 30-60°, particularly 45°. Thus, both of the porous filter plates 2c, 2d are sufficiently exposed

to an activating light.

[0043] Fig. 7 shows a three-dimensional, photocatalytic filter apparatus according to a still further embodiment having three groups of porous filter plates 2e, 2f, 2g; porous filter plates 2e, 2f being assembled in a lattice structure, and oblique porous filter plates 2g crossing the porous filter plates 2e, 2f. Light sources (not shown) are disposed on both sides of the porous filter plates 2e, 2f, 2g. As is clear from Fig. 7, all of the porous filter plates 2e, 2f, 2g are inclined relative to the fluid stream F and an activating light (not shown). The inclination angles  $\theta_9$  is typically 45°. Thus, all of the porous filter plates 2e, 2f, 2g are sufficiently exposed to an activating light.

[0044] Fig. 8 schematically shows a three-dimensional, photocatalytic filter apparatus according to a still further embodiment having a plurality of porous filter plates 2i arranged around a light source 4 and spirally extending along the axis of the light source 4. The porous filter plates 2i are contained in a cylindrical pipe 5. Each porous filter plate 2i is inclined by an angle of 30°, for example, relative to the radial direction of the light source 4 (the activating light direction).

[0045] Fig. 9 shows a three-dimensional, photocatalytic filter apparatus 10 according to a still further embodiment, and Fig. 10 shows a photocatalytic filter unit 8 contained in the three-dimensional, photocatalytic filter apparatus 10 of Fig. 9. Fig. 11 shows a support member 9 contained in the photocatalytic filter unit 8 of

Fig. 10, and Fig. 12 schematically shows the inclination of porous filter plates 2j in the photocatalytic filter unit 8.

[0046] Referring to Fig. 9, the three-dimensional, photocatalytic filter apparatus 10 comprises a cylindrical pipe 5 having an inlet 5a and an outlet 5b for a fluid stream F, a light source 7 positioned at a center of the cylindrical pipe 5, and a three-dimensional, photocatalytic filter unit 8 disposed between the cylindrical pipe 5 and the light source 7. Both ends of the light source 7 are supported by sockets 6, 6' fixed to the cylindrical pipe 5. The light source 7 may be a lamp having a wavelength of 360-390 nm, such as a black light, a fluorescent light, etc. A fan 15 is mounted inside the cylindrical pipe 5 upstream of the three-dimensional, photocatalytic filter unit 8 to generate a fluid stream F.

[0047] Referring to Fig. 10, the three-dimensional, photocatalytic filter unit 8 is constituted by three daisy wheel-shaped, support members 9 and a plurality of porous filter plates 2j supported by the support members 9. The porous filter plates 2j spirally extend along the axis of the light source 7 with inclination relative to the radial direction of the light source 7.

[0048] Each support member 9 is constituted by an annular frame 91 and a plurality of petal portions 92 projecting from the annular frame 91. Each petal portion 92 is inclined by an angle of  $\theta_{10}$  relative to the radial direction X of the light source 7. Each petal portion 92 has a radial groove or slit 92a for receiving an end of each porous filter plate 2j.

[0049] The radial inclination angle  $\theta_{10}$  is preferably determined such that the maximum exposure of each porous filter plate 2j to the light source 7 is achieved. The maximum radial inclination angle  $\theta_{10}$  can geometrically be determined by the number of the porous filter plates 2j and the width (radial length) of each porous filter plate 2j. A typical radial inclination angle  $\theta_{10}$  is 30° for 24 porous filter plates 2j.

[0050] Referring to Fig. 12, the larger the longitudinal inclination angle  $\theta_{11}$ , the likelier the fluid stream F comes to contact with the photocatalyst layer of each porous filter plate 2j. However, too large longitudinal inclination angle  $\theta_{11}$  makes the working of the porous filter plates 2j difficult. For example, the longitudinal inclination angle  $\theta_{11}$  is preferably 15-45° for 24 porous filter plates 2j.

[0051] Fig. 13 shows another support member 90. The support member 90 comprises a disc frame 95, and a plurality of radial projections 96 with a gap 97 between adjacent radial projections 96. An end of the porous filter plate 2j is supported by each gap 97. In this example, the support member 90 has 24 gaps 97 for supporting 24 porous filter plates 2j.

[0052] Returning to Figs. 9-12, pollutants in the fluid stream F such as an exhaust gas, etc. passing through the cylindrical pipe 5 come into contact with the porous filter plates 2j and are oxidatively decomposed by the semiconductor photocatalyst activated by activating light from the light source 7. Because each porous filter plate 2j is inclined relative to the light source 7, the photocatalyst layer can be uniformly and fully irradiated by ultraviolet, thereby ensuring the uniform activation of the semiconductor photocatalyst.

[0053] When nitrogen compounds, sulfur compounds, chlorine compounds, etc. are oxidatively decomposed by the photocatalyst, intermediate products adhere to a surface of the photocatalyst layer. In this case, the three-dimensional, photocatalytic filter unit 8 is taken out of the cylindrical pipe 5 and washed with water, weak acidic water, etc.

[0054] In the third example of the present invention, there is provided a three-dimensional, photocatalytic filter apparatus constituted by a porous filter plate providing a sufficient contact of a fluid stream with a semiconductor photocatalyst, constituted by a porous substrate such as a wire net or mesh, and a porous layer constituted by sintered fine metal particles, and a photocatalyst layer; a support member supporting a light source; and a pipe through which a fluid stream flows in an axial direction. The porous filter plate and the support member are placed in the pipe, and the porous filter plate is fixed around the support member spirally.

[0055] Fig. 14 shows a three-dimensional, photocatalytic filter apparatus comprising a plurality of frustoconical filter plates 2h arranged around a light source 4 and supported by support members (not shown). The photocatalytic surface of each frustoconical filter plate 2h is inclined relative to the fluid stream F by an angle of

5 preferably 15-75°, more preferably 30-60°, particularly 45°.

[0056] Fig. 15 shows a three-dimensional, photocatalytic filter apparatus 100 having a three-dimensional, photocatalytic filter unit 108 comprising a plurality of spiral porous filter plates 2k fixed to a support member 109 disposed around a light source 7. The spiral porous filter plates 2k are inclined relative to the fluid stream F and an activating light emitted from the light source 7 both by an angle of preferably 15-75°, more preferably 30-60°, particularly 45°.

[0057] Pollutants in a fluid stream such as an waste water, etc. penetrating into the cylindrical pipe 5 comes into contact with the porous filter plate 2k, and is oxidatively decomposed by semiconductor photocatalysts activated by activating light from a light source 7, and then the cleaned air is discharged.

[0058] The pipe is preferably made of a material reflecting activating light irradiated from the light source. In the case of illuminating a near ultraviolet ray, the pipe is preferably made of an Al alloy, a stainless steel, etc. Additionally, it is preferred that a photocatalyst layer is formed on the inner surface of the pipe to increase decomposition-efficiency. The pipe preferably has a window with a light transmission properties.

[0059] The above-mentioned, three-dimensional, photocatalytic filter apparatus and the three-dimensional, photocatalytic filter apparatuses may be fixed, vibrated or rotated in a fluid stream. With respect to the three-dimensional, photocatalytic filter apparatus and the three-dimensional, photocatalytic filter apparatus of the present invention, the fluid stream is preferably introduced in a longitudinal direction.

[0060] As described above in detail, a three-dimensional, photocatalytic filter apparatus of the present invention is porous with a large photocatalytic surface area, thereby ensuring enough contact between pollutants in the fluid steam and the semiconductor photocatalyst. It may have a structure from which intermediate products attached thereto are easily removed.

[0061] A three-dimensional, photocatalytic filter apparatus of the present invention is constituted by a porous, photocatalytic filter providing a sufficient contact of a fluid stream with a semiconductor photocatalyst and a light source placed therein. It may have a compact structure, and ensure adequate irradiation over the entire photocatalytic surface to efficiency activate the semiconductor photocatalyst, and make good use of scattered light.

## 50 Claims

1. A three-dimensional, photocatalytic filter apparatus comprising a plurality of porous filter plates each carrying a photocatalyst for de-composing pollutants in a fluid stream which comes into contact with said filter plates, said filter plates being arranged in a path of said fluid stream such that each filter plate

is inclined relative to both said fluid stream and a light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source.

2. The apparatus of claim 1, wherein said flow path means is constituted by a pair of parallel, transparent sheets, and said light source is disposed outside at least one of said sheets.

3. The apparatus of any preceding claim, wherein each of said filter plates is inclined by 15 to 75° relative to said fluid stream and by 15 to 75° relative to said light source.

4. The apparatus of claim 1 or 2, wherein said filter plates intersect each other in a lattice pattern, such that all of them are inclined relative to both said fluid stream and said light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source.

5. The apparatus of claim 4, wherein said filter plates are inclined in one direction by 15 to 75° relative to said light source in the other direction by 75 to 15° relative to said light source.

6. A three-dimensional, photocatalytic filter apparatus comprising

- (a) a flow path means through which a fluid stream containing pollutants passes;
- (b) a plurality of porous filter plates each carrying a photo-catalyst for decomposing said pollutants;
- (c) a light source; and
- (d) at least one support member for arranging each filter plate in a path of said fluid stream in said flow path means at such a three-dimensional position that each filter plate is inclined relative to both said fluid stream and said light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source.

7. The apparatus of claim 6, wherein said flow path means is a pipe having a longitudinal axis along which an elongated light source extends, and a plurality of said filter plates extend in said pipe spirally around said light source in parallel therewith, said filter plates being fixed to said support member such that each of them is inclined relative to both the axial and radial directions of said flow path means.

8. The apparatus of claim 6 or 7, wherein all filter plates extend spirally at an angle of 15 to 75° relative to the axial direction of said light source and are inclined at an angle of 15 to 75° relative to the radial direction of said light source.

9. The apparatus of claim 7 or 8, wherein said pipe is made of a material reflecting an activating light emanating from said light source and has a photocatalyst layer on its inner surface.

10. The apparatus of any one of claims 6 to 9, wherein said support member is an integral daisy-wheel shaped member having an annular frame and a plurality of radially inclined petal portions projecting from said frame, each of said petal portions having a slit or groove for supporting an end of each filter plate.

11. The apparatus of any one of claims 6 to 9, wherein said support member is a disc frame having a plurality of radially inclined slits or grooves each supporting an end of each filter plate.

12. A three-dimensional, photocatalytic filter apparatus comprising

- (a) a flow path means through which a fluid stream containing pollutants passes;
- (b) a plurality of frustoconical porous filter plates each carrying a photocatalyst for decomposing said pollutants;
- (c) an elongated light source extending along the axis of said flow path means; and
- (d) a support member disposed round said light source for arranging said filter plates longitudinally in said flow path means, whereby a conical surface of each filter plate is inclined relative to both said fluid stream and said light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source.

13. A three-dimensional, photocatalytic filter apparatus comprising

- (a) a flow path means through which a fluid stream containing pollutants passes;
- (b) a corrugated porous filter plate carrying a photocatalyst for decomposing pollutants and disposed in said flow path means; and
- (c) a light source disposed outside at least one side of said flow path means, whereby said filter plate is inclined relative to both said fluid stream and said light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source.

14. The apparatus of claim 13, wherein said flow path means is constituted by a pair of parallel, transparent sheets, and said light source is disposed outside at least one side of said flow path means.

side at least one of said sheets.

15. The apparatus of claim 13 or 14, wherein each part of said corrugated filter plate is inclined by 15 to 75° relative to said fluid stream and by 15 to 75° relative to said light source. 5

16. A three-dimensional, photocatalytic filter apparatus comprising

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(a) a flow path means through which a fluid stream containing pollutants passes;  
(b) a spiral porous filter plate carrying a photocatalyst for decomposing said pollutants;  
(c) an elongated light source extending along the axis of said flow path means; and  
(d) a support member disposed around said light source for arranging said spiral filter plate longitudinally in said flow path means, whereby a surface of said filter plate is inclined relative to both said fluid stream and said light source, to achieve sufficient contact with said pollutants and sufficient exposure to said light source. 15

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17. The apparatus of claim 12 or 16, wherein said flow path means is a pipe having a longitudinal axis along which said elongated light source extends. 25

18. The apparatus of any one of claims 16 or 17, wherein a surface of said filter plate is inclined by 15 to 75° relative to the axial direction of said light source and by 15 to 75° relative to the radial direction of said light source. 30

19. The apparatus of any one of claims 1, 6, 7, 12, 16 and 17, wherein the or each filter plate is constituted by a porous substrate, a porous layer of fine particles deposited on at least one surface of said substrate, and a photocatalyst layer deposited on a surface of said porous layer. 35

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20. The apparatus of any preceding claim, wherein said substrate and/or said porous layer is made of Fe, Al, Ti, Cu, alloys of these metals or stainless steel. 45

21. The apparatus of any one of claims 1 to 19, wherein said substrate is constituted by a net or mesh of metal wires or a perforated metal sheet.

22. A photocatalytic filter plate for removing pollutants in a fluid stream, said filter plate comprising a porous substrate, a porous layer of fine particles deposited on at least one surface of said substrate, and a photocatalyst layer deposited on a surface of said porous layer for decomposing said pollutants. 50

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Fig. 1(a)

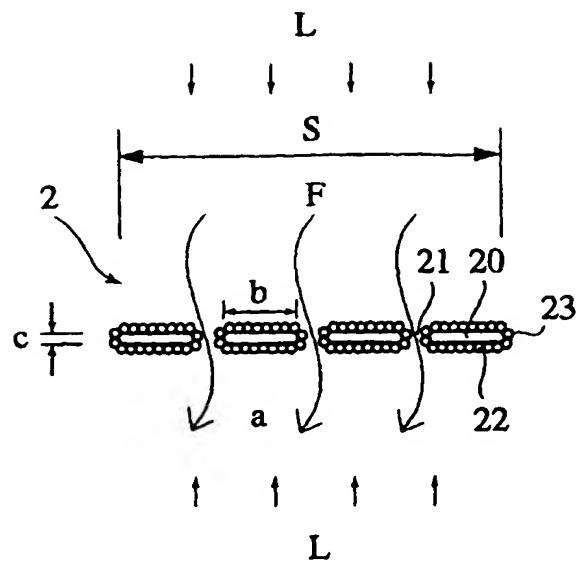


Fig. 1(b)

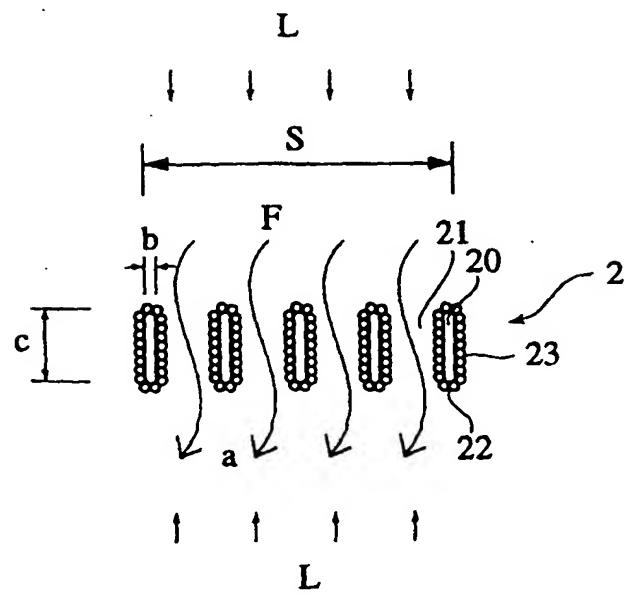


Fig. 2

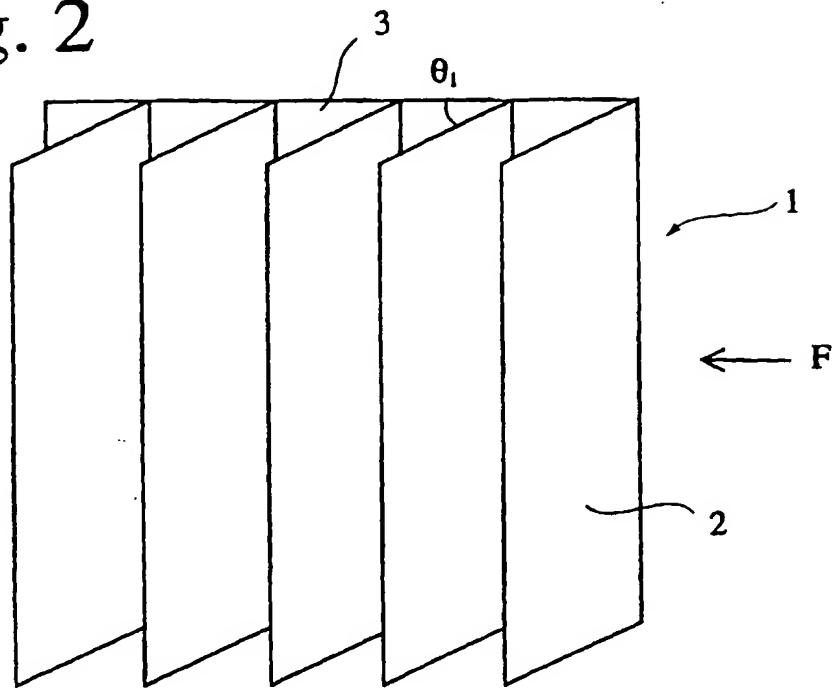


Fig. 3

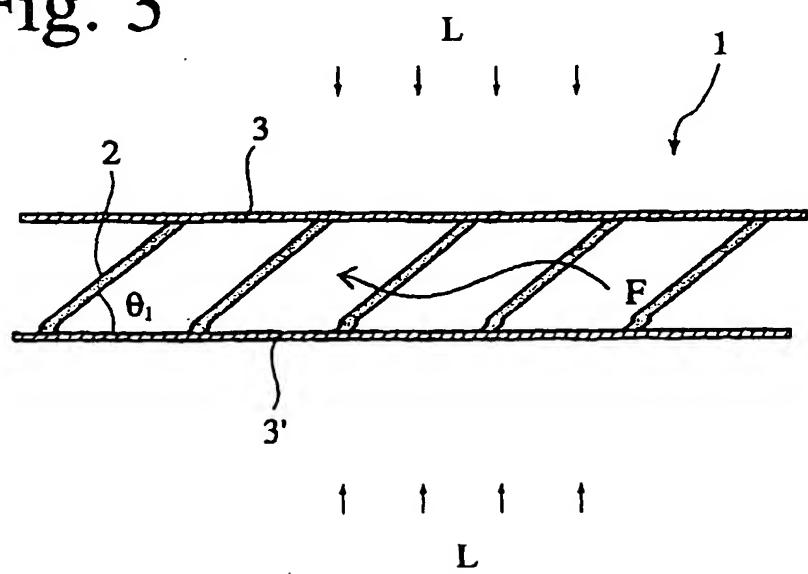


Fig. 4

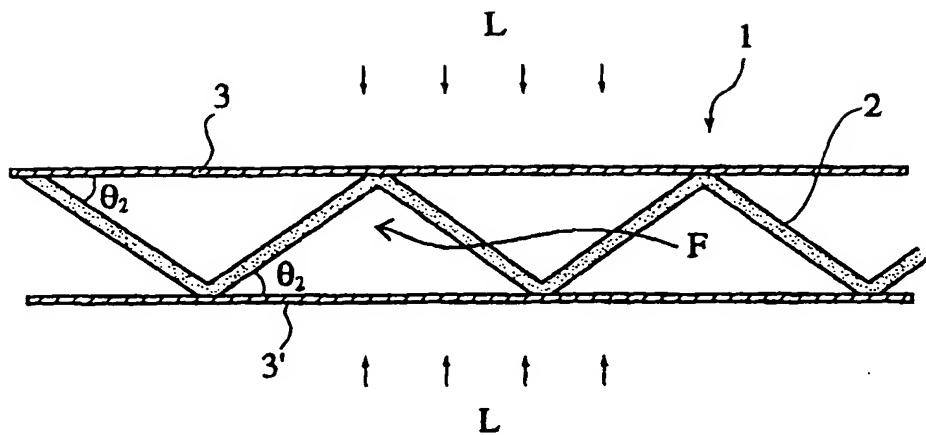


Fig. 7

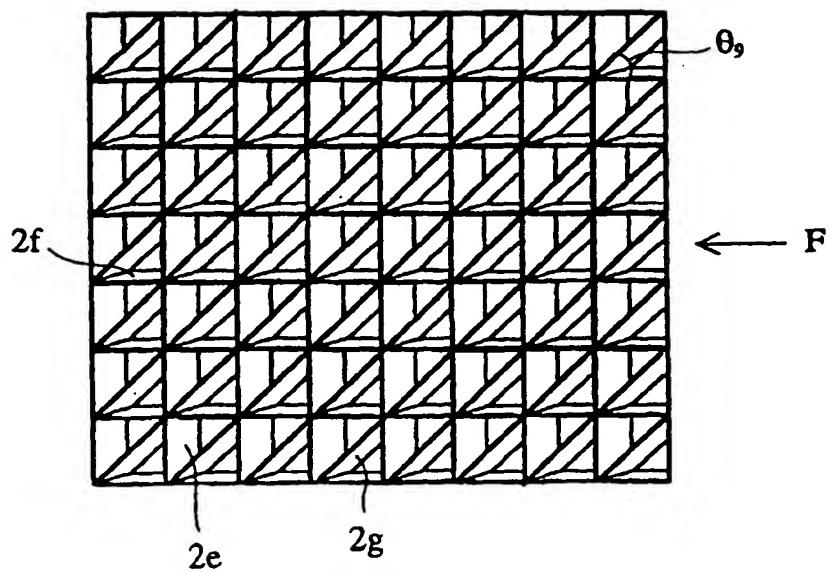


Fig. 5 (a)

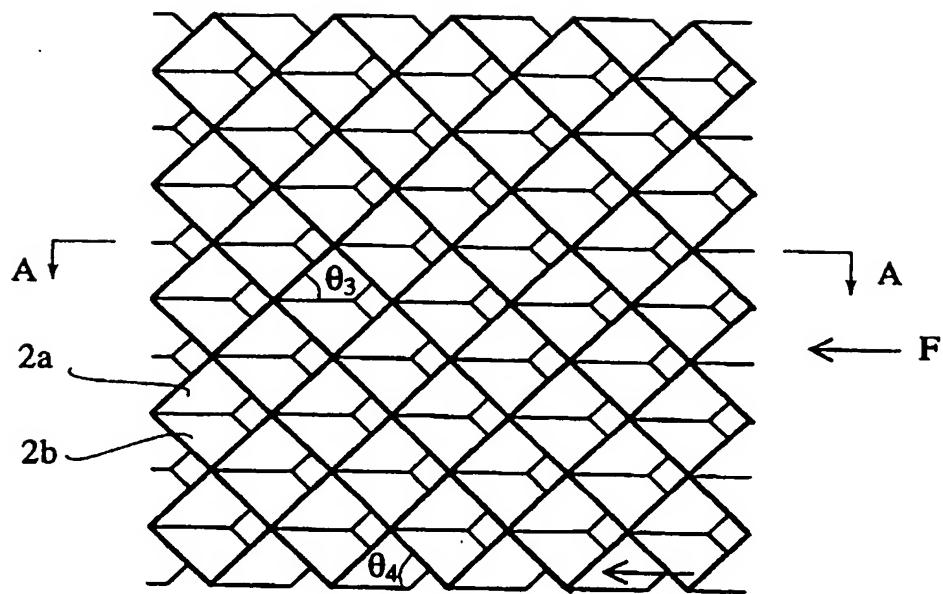


Fig. 5 (b)

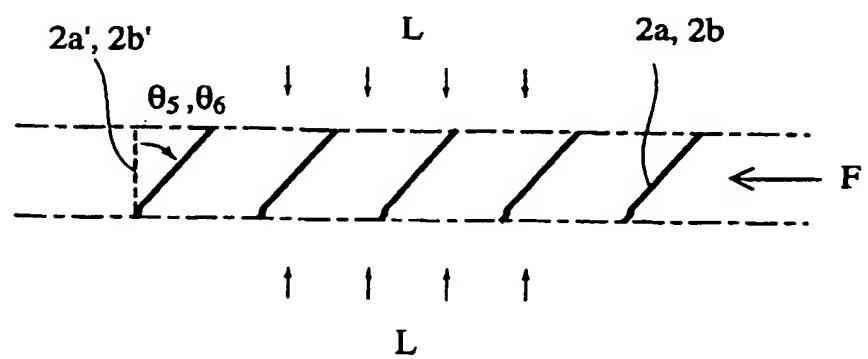


Fig.6

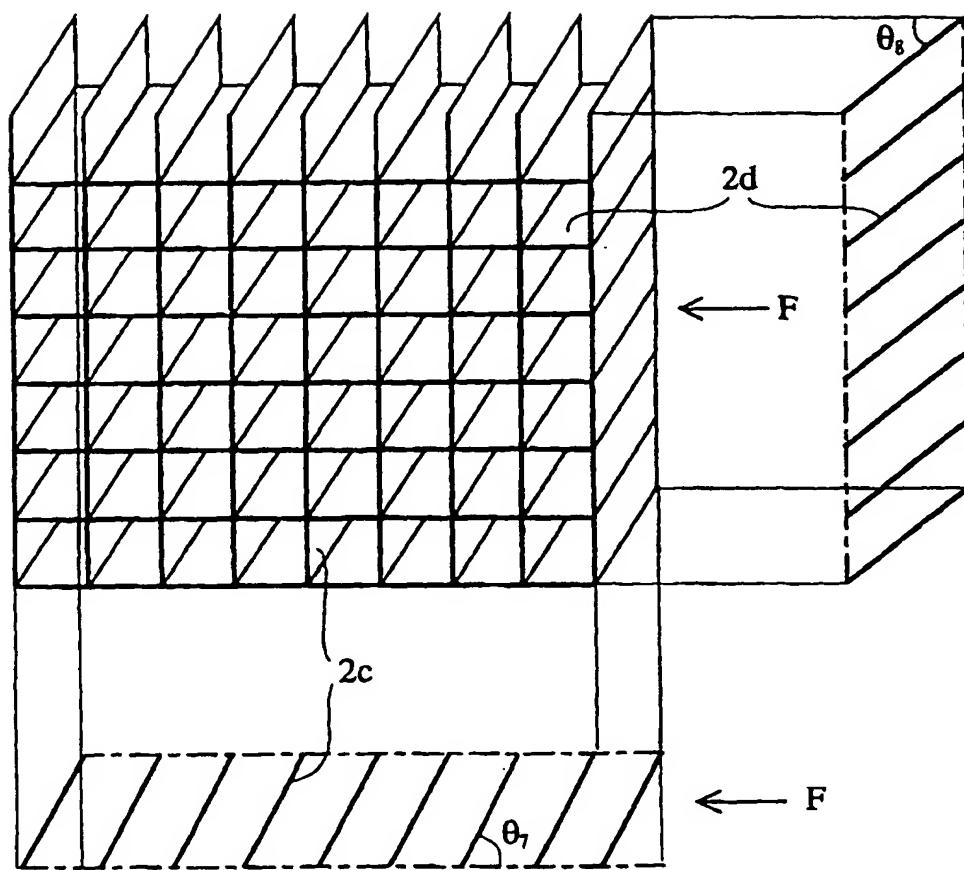


Fig.8

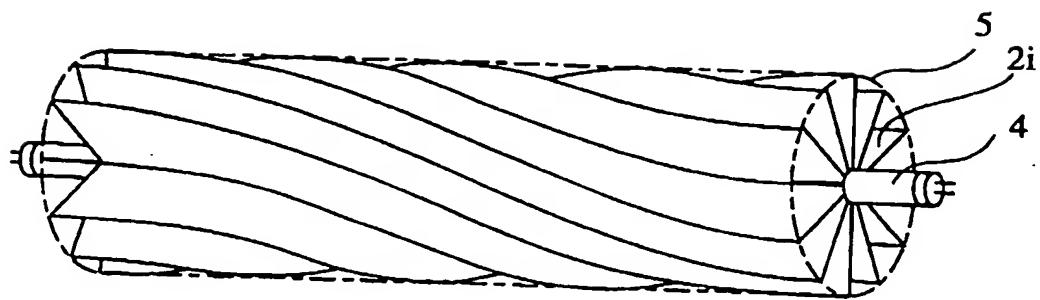


Fig. 14

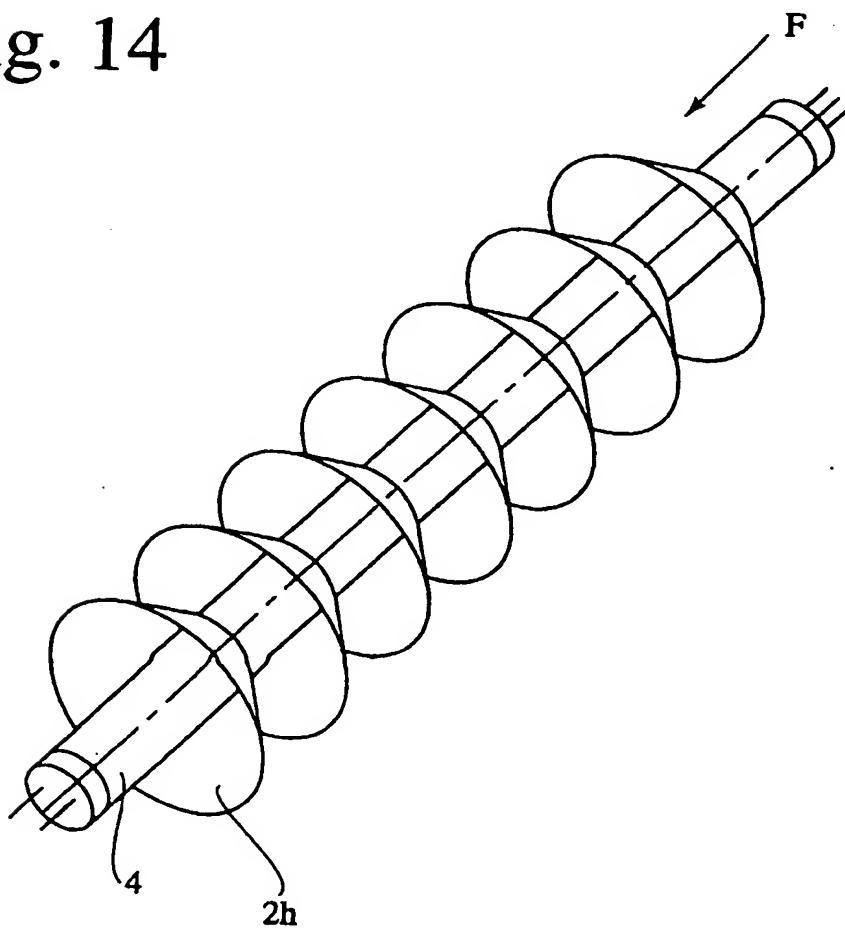


Fig. 9

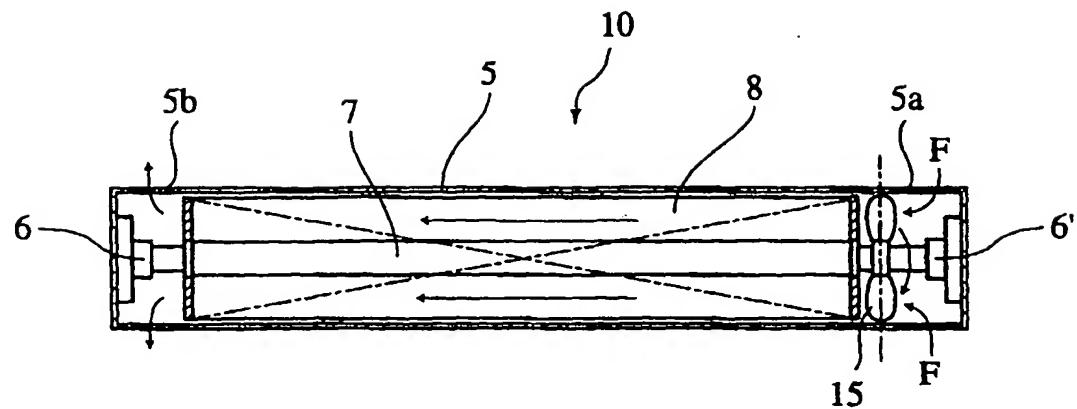


Fig. 10

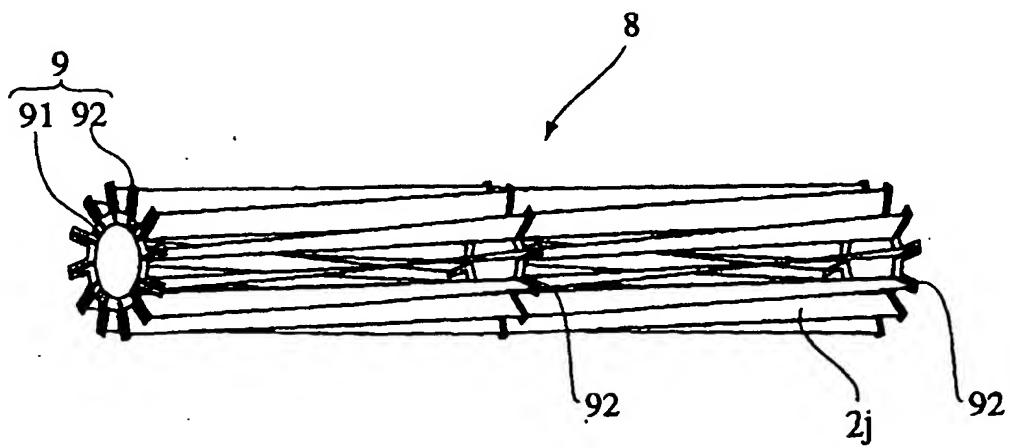


Fig. 11

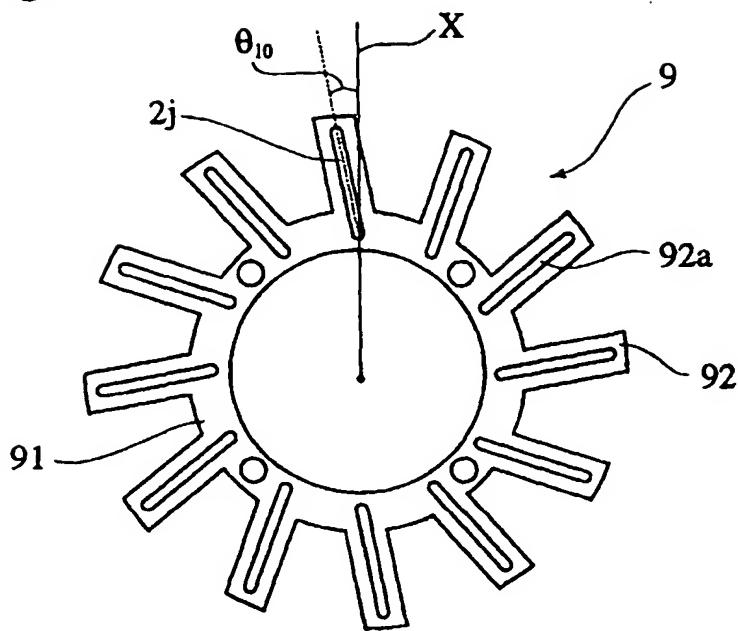


Fig. 12

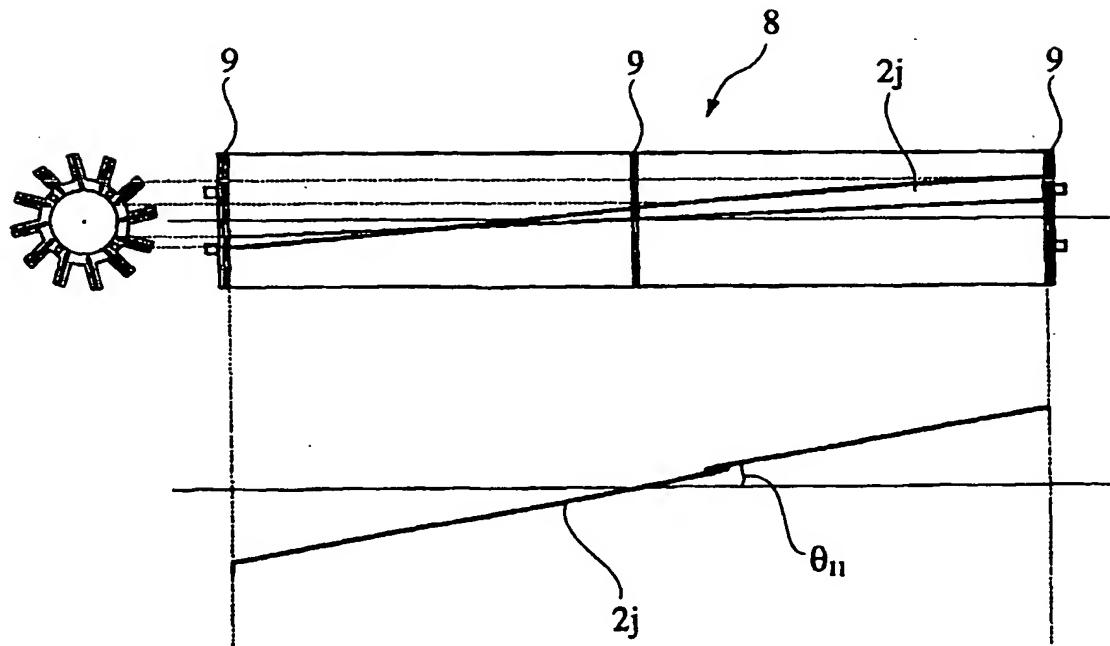


Fig. 13

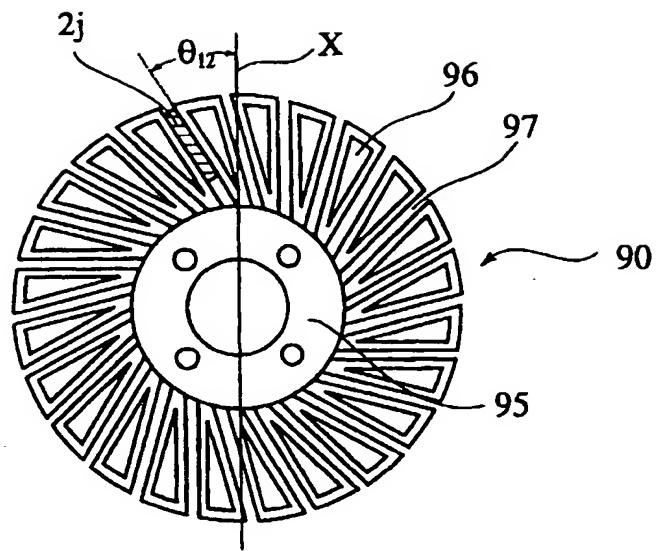
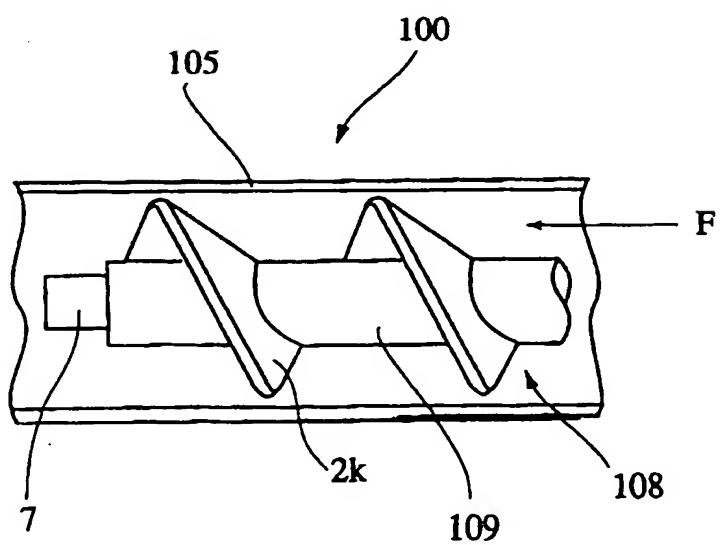


Fig. 15





DOCUMENTS CONSIDERED TO BE RELEVANT			
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	CLASSIFICATION OF THE APPLICATION (Int.Cl.)
X	WO 96 37281 A (MINNESOTA MINING & MFG) 28 November 1996 (1996-11-28)  * figures 1-9 *	1-9, 13-15, 19-22	B01D53/88 B01D53/00 C02F1/30 C02F1/72 B01J19/12
X	WO 96 00190 A (ARTHUR RONALD WILLIAM ; BOYDEN BRACE HARVEY (AU)) 4 January 1996 (1996-01-04) * page 2, line 14 - line 20; figure 2 *	1-9, 13-15, 19-22	
X	WO 97 40936 A (ZENTOX CORP) 6 November 1997 (1997-11-06)  * claims; figures 2,3 *	1-9, 13-15, 19-22	
X	EP 0 826 531 A (EQUOS RESEARCH KK ; AISIN AW CO (JP); FUJISHIMA AKIRA (JP); HASHIMO) 4 March 1998 (1998-03-04) * figures 3,5 *	1-9, 13-15, 19-22	
X	US 5 516 492 A (BERMAN ELLIOT ET AL) 14 May 1996 (1996-05-14) * figures *	1-9, 12, 16-22	TECHNICAL FIELDS SEARCHED (Int.Cl.)
X	US 5 069 885 A (RITCHIE DAVID G) 3 December 1991 (1991-12-03) * figures 1-5 *	1-11, 16-22	B01D C02F B01J
X,D	PATENT ABSTRACTS OF JAPAN Vol. 1997, no. 11, 28 November 1997 (1997-11-28) & JP 09 187491 A (MATSUSHITA ELECTRIC WORKS LTD), 22 July 1997 (1997-07-22) * abstract; figures *	1,6,22  -/-	
The present search report has been drawn up for all claims			
Place of search	Date of completion of the search	Examiner	
MUNICH	22 February 2000	Eijkelenboom, A	
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons  & : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			



European Patent  
Office

## EUROPEAN SEARCH REPORT

Application Number  
EP 99 11 8480

DOCUMENTS CONSIDERED TO BE RELEVANT			CLASSIFICATION OF THE APPLICATION (Int.Cl.)
Category	Citation of document with indication, where appropriate, of relevant passages	Relevant to claim	
X, D	<p><b>PATENT ABSTRACTS OF JAPAN</b>            Vol. 1995, no. 07,            31 August 1995 (1995-08-31)            &amp; JP 07 108138 A (AGENCY OF IND SCIENCE &amp;            TECHNOL; OTHERS: 01),            25 April 1995 (1995-04-25)            * abstract; figure *</p> <hr/>	1,6,22	
			TECHNICAL FIELDS SEARCHED (Int.Cl.)
<p>The present search report has been drawn up for all claims</p>			
Place of search	Date of completion of the search		Examiner
MUNICH	22 February 2000		Eijkenboom, A
CATEGORY OF CITED DOCUMENTS		T : theory or principle underlying the invention E : earlier patent document, but published on, or after the filing date D : document cited in the application L : document cited for other reasons A : member of the same patent family, corresponding document	
X : particularly relevant if taken alone Y : particularly relevant if combined with another document of the same category A : technological background O : non-written disclosure P : intermediate document			

**ANNEX TO THE EUROPEAN SEARCH REPORT  
ON EUROPEAN PATENT APPLICATION NO.**

EP 99 11 8480

This annex lists the patent family members relating to the patent documents cited in the above-mentioned European search report. The members are as contained in the European Patent Office EDP file on. The European Patent Office is in no way liable for these particulars which are merely given for the purpose of information.

22-02-2000

Patent document cited in search report		Publication date		Patent family member(s)		Publication date
WO 9637281	A	28-11-1996		CA 2150320 A AU 5675096 A CN 1185755 A EP 0828551 A JP 11505746 T		27-11-1996 11-12-1996 24-06-1998 18-03-1998 25-05-1999
WO 9600190	A	04-01-1996		AU 709825 B AU 2779795 A CA 2193643 A		09-09-1999 19-01-1996 04-01-1996
WO 9740936	A	06-11-1997		US 5766455 A AU 2829897 A CA 2253706 A DE 19781738 T GB 2328384 A		16-06-1998 19-11-1997 06-11-1997 12-05-1999 24-02-1999
EP 0826531	A	04-03-1998		JP 10071323 A US 5948355 A		17-03-1998 07-09-1999
US 5516492	A	14-05-1996		NONE		
US 5069885	A	03-12-1991		CA 2040447 A		24-10-1991
JP 09187491	A	22-07-1997		NONE		
JP 07108138	A	25-04-1995		JP 2777623 B		23-07-1998